

**IN THE UNITED STATES PATENT AND TRADEMARK OFFICE**

In re Application of: James B. Kimble, Charles A. Drake, Jianhua Yao and  
An-hsiang Wu

Serial No.: 08/956,082

Group Art Unit: 1764

Filed: October 23, 1997

Examiner: W. Griffin

For: CATALYST COMPOSITION COMPRISING ZINC COMPOUND OR  
BORON COMPOUND AND HYDROCARBON CONVERSION PROCESS

**DECLARATION UNDER 37 CFR 1.131**

Assistant Commissioner for Patents  
Washington, D.C. 20231

Sir:

We, James B. Kimble, Charles A. Drake, Jianhua Yao and

An-hsiang Wu, do declare and say:

that we are the undersigned inventors of application Ser. No. 08/956,082,  
filed October 23, 1997;

that USPN 5,883,034 substantially shows but does not claim the same  
patentable invention as set out in the claims of application Ser. No. 08/956,082,  
filed October 23, 1997;

that the invention set out in application Ser. No. 08/956,082 was completed  
in this country before July 9, 1997, the effective filing date of application Ser. No.  
890,540 which is the parent of the C.I.P. application that matured into USPN  
5,883,034;

that the filing date of the present application, October 23, 1997, is less than a year after the filing of Ser. No. 890,540;

that attached hereto as Exhibit A is a "Patent Idea Record" of Phillips Petroleum Company (dates removed) signed by James B. Kimble, Charles A. Drake and Jianhua Yao showing the completion of part of the invention set out and claimed in application Ser. No. 08/956,082;

that attached hereto as Exhibit B is a "Patent Idea Record" of Phillips Petroleum Company (dates removed) signed by Charles A. Drake and An-hsiang Wu showing the completion of part of the invention set out and claimed in application Ser. No. 08/956,082;

that attached hereto as Exhibit C is a page from a laboratory notebook (dates removed) signed by Charles A. Drake verifying the steam treatment of the catalyst 37800-80-2 disclosed in Exhibit B;

that all the pertinent dates which have been removed from Exhibits A, B and C are prior to July 9, 1997;

further Declarants saith not;

We hereby declare that all statements made herein of our individual knowledge are true and that all statements made on information and belief are believed to be true; further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or

imprisonment, or both, under 18 U.S.C. 1001 and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

James B Kimble  
James B. Kimble

10/20/99  
Date

Charles A Drake  
Charles A. Drake

10/25/99  
Date

Jianhua Yao  
Jianhua Yao

10/21/99  
Date

An-hsiang Wu  
An-hsiang Wu

10/21/99  
Date

9785

PATENT IDEA RECORD  
PHILLIPS PETROLEUM COMPANY & SUBSIDIARIES

PATENT  
DIVISION

EXHIBIT A

SUBJECT Zinc compound promoted ZSM-5 catalyst

Please identify earlier recordings and/or discussions of this invention which you can recall: Drawing(s) \_\_\_\_\_ Notebook (or Diary) 37158-46-2

Correspondence \_\_\_\_\_ Work (or purchase) order(s) \_\_\_\_\_ Discussions \_\_\_\_\_

Briefly describe the idea. Set forth its application, operation and novel features. A freehand sketch of the idea will be appreciated.

Is this patent idea in use? \_\_\_\_\_ If so, date of first usage \_\_\_\_\_ If not, are there any plans to use it? \_\_\_\_\_

If the patent idea is a solution to a problem, please state the problem, describe the idea, and indicate how the idea solves the problem.

In the converting hydrocarbon to BTX and olefins ( $C_2^+$  +  $C_3^+$ ) process, better economics is expected when the BTX + olefins yield and olefins/BTX yield ratio are maximized. It is also desired that the coke formation on the catalyst is very low in order to maintain the stability of catalyst. It is now found that by mixing ZSM-5 powder with Zinc compound powder (e.g.  $ZnAl_2O_4$ ,  $ZnTiO_3$ ,  $ZnSiO_4$ ) and embedding them into  $SiO_2$  matrix, then steaming the catalyst at 600°C for 6 hr, the resulting catalysts showed higher BTX + olefin yield, higher Product value<sup>(PV)</sup> over gasoline, and much lower coke formation than commercial ZSM-5 catalyst. The example is shown below.

Run #	Cat ID	Cat Description	Olefin + BTX (wt %)	Olefin/BTX	PV over gasoline (lb)	Coke formation (%/hr)
37158-46-2	Zeocat	Commercial ZSM-5 catalyst	58.0	0.36	2.1	4.7
37158-79-2	37157-16-2	PZ/SOH + $ZnTiO_3$ + $SiO_2$ binder + 6hr steam	64.6	0.43	3.2	0.2
37158-84-2	37157-23-3	PZ/SOH + $Zn_2SiO_4$ + $SiO_2$ binder + 6hr steam	60.8	0.51	3.1	0.2
37158-84-1	37157-23-2	PZ/SOH + $ZnAl_2O_4$ + $SiO_2$ binder + 6hr steam	58.6	0.55	3.1	0.3
37158-82-2	37157-21-3	PZ/SOH + $ZnTiO_3$ + $Al_2O_3$ binder + 6hr steam	60.0	0.50	3.0	1.5

WITNESS: PZ/SOH: ZSM-5 powder

READ AND UNDERSTOOD BY:

John W. Miller

DATE

PLEASE SIGN AND DATE, AS INDICATED, AND SUBMIT TO PATENT DIVISION.

9296

PAI

J B Kinnale 427494 DATE \_\_\_\_\_  
(ONE GIVEN NAME/ONE INITIAL/LAST NAME) (EMPLOYEE NO.)  
370A PL BUSINESS MAILING ADDRESS Biville CITY GROUP/STAFF/SUBSIDIARY R & D

PAI

Charles A. Decker 232581 DATE \_\_\_\_\_  
(ONE GIVEN NAME/ONE INITIAL/LAST NAME) (EMPLOYEE NO.)

PAI

322 PL BUSINESS MAILING ADDRESS CITY GROUP/STAFF/SUBSIDIARY  
John W. Miller 503328 DATE \_\_\_\_\_  
(ONE GIVEN NAME/ONE INITIAL/LAST NAME) (EMPLOYEE NO.)  
370 PL Bartsleville R & D

PATENT IDEA RECORD  
PHILLIPS PETROLEUM COMPANY & SUBSIDIARIES

PATENT  
DIVISION

SUBJECT Improved Aromatization Catalyst

EXHIBIT B

Please identify earlier recordings and/or discussions of this invention which you can recall: Drawing(s) \_\_\_\_\_ Notebook(s) \_\_\_\_\_

Correspondence \_\_\_\_\_ Work (or purchase) order(s) \_\_\_\_\_ Discussions \_\_\_\_\_

Briefly describe the idea. Set forth its application, operation and novel features. A freehand sketch of the idea will be appreciated.

Is this patent idea in use? \_\_\_\_\_ If so, date of first usage \_\_\_\_\_ If not, are there any plans to use it? \_\_\_\_\_

If the patent idea is a solution to a problem, please state the problem, describe the idea, and indicate how the idea solves the problem.

One of the goals of the GCL process is to maximize the yield of BTX while minimizing the amount of coke. It has previously been shown that incorporation of zinc silicate into a mixture of ZSM-5 and binder gives good yields of BTX while producing low amounts of coke. It has now been found that the addition of BrO<sub>3</sub> to such a catalyst gives an even higher yield of BTX and lower coke as shown below.

Run No	Cat No	Cat Description	oefin Yield	BTX Yield	% Coke
1802-40-4	3880-80-1	PZ + Ludox + Zr Silicate	21.3	40.5	0.26
110482-6	37800-80-2	PZ + Ludox + Zr Silicate + BrO <sub>3</sub>	18.8	44.4	0.17

WITNESS:

READ AND UNDERSTOOD BY:

John W. Miller

DATE \_\_\_\_\_

PLEASE SIGN AND DATE, AS INDICATED, AND SUBMIT TO PATENT DIVISION.

19574

SIGNATURE OF INVENTOR(S):

Charles A. Deshe  
(ONE GIVEN NAME/ONE INITIAL/LAST NAME)

232581  
(EMPLOYEE NO.)

DATE \_\_\_\_\_

B2L PL  
BUSINESS MAILING ADDRESS

Bulle  
CITY

GROUP/STAFF/SUBSIDIARY

Am - Henry W.  
(ONE GIVEN NAME/ONE INITIAL/LAST NAME)

742643  
(EMPLOYEE NO.)

DATE \_\_\_\_\_

333A PL  
BUSINESS MAILING ADDRESS

CITY

GROUP/STAFF/SUBSIDIARY

(ONE GIVEN NAME/ONE INITIAL/LAST NAME)

(EMPLOYEE NO.)

DATE \_\_\_\_\_

BUSINESS MAILING ADDRESS

CITY

GROUP/STAFF/SUBSIDIARY

1/2 Catalyst Prep -

1800-80-1

EXHIBIT C

Prepare same mixture of 20g Zeocat,  
0.4g zinc ortho silicate and enough  
ludox to make paste - ~~effluent~~ <sup>granulate</sup> dry -  
Steam 650C / 4 hrs - 16 ml / 4 hrs

1800-80-2 Catalyst #

Prepare mixture of 20g Zeocat, 0.4g zinc  
ortho silicate, 0.4g barium oil and enough  
ludox to make paste - ~~effluent~~ <sup>granulate</sup> dry -  
Steam 650C / 4 hrs - 28 ml / 4 hrs

1800-80-3 - Prepare mixture of 20g Zeocat  
powder and 5g silica gel 4-20  $\mu$  -  
add H<sub>2</sub>O to make paste - ~~effluent~~ <sup>granulate</sup> dry -  
calcine 550C / 3 hrs  
20 ml / H<sub>2</sub>O